PATENT ABSTRACTS OF JAPAN

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HOZUMI TAKESHI

(54) INTERLAYER INSULATION ADHESIVE FOR MULTILAYER PRINTED WIRING BOARD AND COPPER FOIL

(57)Abstract:
PROBLEM TO BE SOLVED: To obtain an adhesive having improved flame retardancy in spite of being a nonhalogen adhesive and improved in heat resistance, electrical properties, storage stability and high-temperature curability by including a specified sulfur-component-containing thermoplastic resin, an inorganic filler, an epoxy resin and an epoxy resin curing agent.
SOLUTION: 30-90 wt.% sulfur-component-containing thermoplastic resin having a weight-average molecular weight of 103-105, 10-70 wt.% epoxy resin having an epoxy resin equivalent of 500 or below, 5-50 wt.%, based on component B, inorganic filler and an epoxy resin curing agent having a melting point of 130°C or above are dissolved in a solvent such as acetone, methyl ethyl ketone, toluene or xylene to obtain an insulation adhesive for a multilayer printed wiring board. An interlayer-insulation-adhesive-coated copper foil obtained by applying the adhesive in a thickness of 15-120 μm to the anchor surface of a copper foil and drying the adhesive at 80-130°C is laminated on an inner layer circuit board by means of a dry film laminator, and the adhesive is cured to obtain a multilayer printed wiring board having an outer layer circuit.

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30-90% PES
5-50% silica
Ex 1 PES (5003P) MW 24,000
nov ep
bis A ep
Bu SO4
Cx. not bis F eps

AN 1999:238657 CAPLUS
DN 130:312940
ED Entered STN: 19 Apr 1999
TI Interlayer insulating epoxy adhesives for multilayer printed circuit boards and their coated copper foils
IN Kamisaka, Masao; Hozumi, Takeshi
PA Sumitomo Bakelite Co., Ltd., Japan
SO Jpn. Kokai Tokkyo Koho, 6 pp.
CODEN: JKXXAF
DT Patent
LA Japanese
IC ICM C09J163-00
ICS C09J181-00; H05K003-38; H05K003-46; B32B027-38; C08L063-00
CC 38-3 (Plastics Fabrication and Uses)
Section cross-reference(s): 76
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	JP 2002327161	A2	20021115	JP 2002-74368	19970926
PRAI	JP 1997-262425	A3	19970926		

CLASS

PATENT NO.	CLASS	PATENT FAMILY CLASSIFICATION CODES
JP 11100562	ICM	C09J163-00
	ICS	C09J181-00; H05K003-38; H05K003-46; B32B027-38; C08L063-00
	IPCI	C09J0163-00 [ICM,6]; C09J0181-00 [ICS,6]; H05K0003-38 [ICS,6]; H05K0003-46 [ICS,6]; B32B0027-38 [ICS,6]; C08L0063-00 [ICS,6] <--
JP 2002327161	IPCI	C09J0163-00 [ICM,7]; C09J0009-00 [ICS,7]; C09J0185-00 [ICS,7]; H05K0003-46 [ICS,7]

AB The adhesives contain S-containing thermoplastics with Mw 103 - 105, inorg. fillers, epoxy resins with epoxy equivalent ≤500, and curing agents for the epoxy resins. Thus, Cu foil was applied with an adhesive varnish containing 5003P (OH-terminated polyether-polysulfone, Mw 24,000) 100, Epiclon N 770 (novolak epoxy resin, epoxy equivalent 190) 30, Epiclon 830S (bisphenol F-type epoxy resin; epoxy equivalent 175) 15, 2-methylimidazole 5, KR 46B (titanate-type coupling agent) 0.2, and BaSO4 20 parts in Me Et ketone and DMF, and dried. The Cu foil with the adhesive was laminated with an circuit board having an undercoat layer (prepared from 100 parts bisphenol A epoxy resin and 40 parts glycidyl methacrylate) and heat cured. The board showed surface smoothness 5 μm (JIS B 0601), good moisture and solder heat resistance, peeling strength (JIS C 6486) 1.4 kg/cm, and fire resistance (JIS C 6481) V-0.

ST elec insulator adhesive printed circuit board; polyether polysulfone novolak epoxy adhesive copper; bisphenol epoxy polysulfone adhesive heat resistance; multilayer printed circuit board interlayer adhesive; fire resistance epoxy adhesive halogen free

IT Electric insulators
Electric insulators
(adhesives; copper foils coated with interlayer insulating epoxy adhesives for multilayer printed circuit boards)

IT Polythiophenylenes
RL: POF (Polymer in formulation); PRP (Properties); TEM (Technical or engineered material use); USES (Uses)
(adhesives; copper foils coated with interlayer insulating epoxy adhesives for multilayer printed circuit boards)

IT Phenoxy resins
RL: POF (Polymer in formulation); PRP (Properties); TEM (Technical or engineered material use); USES (Uses)
(bisphenol S-based, adhesives; copper foils coated with interlayer insulating epoxy adhesives for multilayer printed circuit boards)

IT Fire-resistant materials

Heat-resistant materials
Semiconductor device fabrication
(copper foils coated with interlayer insulating epoxy adhesives for multilayer printed circuit boards)

IT Adhesives
Adhesives
(dielec.; copper foils coated with interlayer insulating epoxy adhesives for multilayer printed circuit boards)

IT Polysulfones, uses
Polysulfones, uses
Polysulfones, uses
RL: DEV (Device component use); IMF (Industrial manufacture); POF (Polymer in formulation); PRP (Properties); TEM (Technical or engineered material use); PREP (Preparation); USES (Uses)
(epoxy-polyether-; copper foils coated with interlayer insulating epoxy adhesives for multilayer printed circuit boards)

IT Polyethers, uses
Polyethers, uses
Polyethers, uses
RL: DEV (Device component use); IMF (Industrial manufacture); POF (Polymer in formulation); PRP (Properties); TEM (Technical or engineered material use); PREP (Preparation); USES (Uses)
(epoxy-polysulfone-; copper foils coated with interlayer insulating epoxy adhesives for multilayer printed circuit boards)

IT Printed circuit boards
(multilayer; copper foils coated with interlayer insulating epoxy adhesives for multilayer printed circuit boards)

IT Epoxy resins, uses
Epoxy resins, uses
Epoxy resins, uses
RL: DEV (Device component use); IMF (Industrial manufacture); POF (Polymer in formulation); PRP (Properties); TEM (Technical or engineered material use); PREP (Preparation); USES (Uses)
(polyether-polysulfone-; copper foils coated with interlayer insulating epoxy adhesives for multilayer printed circuit boards)

IT 106-89-8DP, Epichlorohydrin, polymers with 5003P, bisphenol F, and Epiclon N 770 1333-16-0DP, polymers with 5003P, epichlorohydrin, and Epiclon N 770 2095-03-6DP, Bisphenol F diglycidyl ether, polymers with polyether sulfones, epoxy resins, and optionally methyltetrahydrophthalic anhydride 25135-51-7DP, Udel P 1700, polymers with BREN-S, Epiclon 830, and methyltetrahydrophthalic anhydride 25667-42-9DP, 5003P, polymers with Epiclon N 770 and bisphenol F-type epoxy resins 26590-20-5DP, Methyltetrahydrophthalic anhydride, polymers with Udel P 1700, BREN-S, and Epiclon 830 93195-67-6DP, BREN-S, polymers with Udel P 1700, Epiclon 830, and methyltetrahydrophthalic anhydride 99241-45-9DP, Epiclon N 770, polymers with 5003P and bisphenol F-type epoxy resins
RL: DEV (Device component use); IMF (Industrial manufacture); POF (Polymer in formulation); PRP (Properties); TEM (Technical or engineered material use); PREP (Preparation); USES (Uses)
(copper foils coated with interlayer insulating epoxy adhesives for multilayer printed circuit boards)

IT 7440-50-8, Copper, uses
RL: DEV (Device component use); PRP (Properties); USES (Uses)
(copper foils coated with interlayer insulating epoxy adhesives for multilayer printed circuit boards)

IT 693-98-1, 2-Methylimidazole
RL: CAT (Catalyst use); USES (Uses)
(crosslinking agent; copper foils coated with interlayer insulating epoxy adhesives for multilayer printed circuit boards)

IT 7727-43-7, Barium sulfate
RL: MOA (Modifier or additive use); USES (Uses)
(filler; copper foils coated with interlayer insulating epoxy adhesives for multilayer printed circuit boards)

DERWENT-ACC-NO: 1999-296656

DERWENT-WEEK: 200330

TITLE: Insulative adhesive layer for multilayered printed circuits - has thermoplastic resin containing sulfur component, inorganic filler, epoxy resin and hardener

PATENT-ASSIGNEE: SUMITOMO BAKELITE CO LTD[SUMB]

PRIORITY-DATA: 1997JP-0262425 (September 26, 1997)

PATENT-FAMILY:

PUB-NO	PUB-DATE	LANGUAGE	PAGES	MAINIPC
JP 11100562 A	April 13, 1999	N/A	006	C09J 163/00

APPLICATION-DATA:

PUB-NO	APPL-DESCRIPTOR	APPL-NO	APPL-DATE
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RELATED-ACC-NO: 2003-304262

ABSTRACTED-PUB-NO: JP 11100562A

BASIC-ABSTRACT:

NOVELTY - The adhesive layer has a thermoplastic resin (containing a sulfur component) of 103-105 weight average molecular weight, an inorganic filler, 500 or less weight equivalents of an epoxy resin and an epoxy resin hardener.

USE - For multilayered printed circuits.

ADVANTAGE - The layer has superior durability, and the fre resistance, heat resistance, electrical property and water resistance of the printed circuit is improved.

CHOSEN-DRAWING: Dwg.0/1

TITLE-TERMS: INSULATE ADHESIVE LAYER MULTILAYER PRINT CIRCUIT THERMOPLASTIC RESIN CONTAIN SULPHUR COMPONENT INORGANIC FILL EPOXY RESIN HARDEN

DERWENT-CLASS: A85 G03 L03 P73 V04

CPI-CODES: A05-A01E2; A08-D01; A08-R01; A12-E07A; G03-B02E2; L03-H04E3;

EPI-CODES: V04-R05A; V04-R07L; V04-R07P1;

SECONDARY-ACC-NO:
CPI Secondary Accession Numbers: C1999-087415
Non-CPI Secondary Accession Numbers: N1999222975

Partial Translation of
Japanese Patent Application No. 11-100562

[0007] In the present invention, the sulfur component containing thermoplastic resin having a weight average molecular weight of 10^3 - 10^5 of (i) component, is blended for the purposes of making a epoxy resin composition in non-halogen flame-resistant, reducing softening of resin at the time of formation and maintaining the thickness in the insulation layer, assigning flexibility to the epoxy resin composition, and making an insulation resin highly heat-resistant. Moreover, the resin is considered to improve electric characteristics. As the sulfur component containing thermoplastic resin having a weight average molecular weight of 10^3 - 10^5 of (i) component, polyphenylene sulfide, polysulfone, polyether sulfone, and bisphenol S-type phenoxy resin are used. The addition rate of the high-molecular-weight sulfur component containing thermoplastic resin is 30 to 90% by weight based on the entire resin which is the total weight with epoxy resin of (ii) component. If the addition rate is smaller than 30 % by weight, flame resistance cannot be sufficiently expressed. On the other hand, if the addition rate is higher than 90 % by weight, flame resistance can be expressed, but adhesive composition becomes stiff and lacks elasticity For this reason, adhesion to an irregular surface of the base material is poor at the time of lamination and forming void is caused. In addition, if the terminal of the sulfur component containing thermoplastic resin is denatured with a hydroxyl group, a carboxyl group or an amino group, its reactivity with epoxy resin is preferable. Thus, phase separation between the sulfur component containing thermoplastic resin and epoxy resin can be restricted after heat curing and the heat resistance of a cured material can be improved. It is therefore desirable that the denaturation should occur

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] This invention relates to the copper foil which coated the layer insulation adhesives for epoxy resin system multilayer printed wiring boards, and it.
[0002]
[Description of the Prior Art] When manufacturing a multilayer printed wiring board conventionally, one or more prepreg sheets which it sank [sheets] into the glass-fabrics base material, and made it carry out semi-hardening of the epoxy resin on the inner layer circuit board in which the circuit was formed were piled up, copper foil was further piled up on it, and it has passed through the process of really [application-of-pressure] fabricating with a hot-platen press. However, in order to make the impregnating resin in prepreg re-flow with heat, to make it harden under a constant pressure, and to make homogeneity carry out hardening shaping, 1 - 1.5 hours is required of this process. Thus, in this top, the production process serves as high cost by the cost of a multilayer laminating press and glass-fabrics prepreg etc. for a long time. In addition, because of the approach of carrying out impregnation of the resin to glass fabrics, the thickness between circuitry layers was restricted by glass fabrics, and pole thinning of the whole multilayer printed wiring board was also difficult.
[0003] In order to solve these problems in recent years, heating pressing by the hot-platen press is not performed, but the technique of the multilayer printed wiring board by the build up method in which glass fabrics are not used for layer insulation material attracts attention anew. In the multilayer printed wiring board by the build up method, when a film-like layer insulation resin layer is used, in order to lose the insulating substrate, circuit, and level difference of a inner layer circuit plate and to graduate the front face, it has become common to apply an under coat agent to a inner layer circuit plate. As this typical example, the under coat agent applied to the inner layer circuit plate laminates the copper foil which carried out the coat of the layer insulation adhesives in un-hardening, semi-hardening, or the condition of having hardened, and a multilayer printed wiring board is obtained by really hardening. By such approach, the lamination of the copper foil which carried out the coat of the layer insulation adhesives since the level difference by the

circuit of a inner layer circuit plate became small is easy, and need's [of taking into consideration the copper foil survival rate of a inner layer circuit plate] decreases.
[0004] In such a process, the layer insulation adhesives by which the coat was carried out to copper foil become soft too much at the time of lamination shaping, and layer insulation thickness cannot be secured. At the time of heat curing, melt viscosity falls too much and a wrinkle occurs. Moreover, the hardening reaction advanced at the time of the preservation, and when laminated to the inner layer circuit plate with which the under coat agent was applied, the problem referred to as that shaping is not really performed good has arisen. Furthermore, although flameproofing is difficult and the trouble accompanying the densification of a multilayer printed wiring board of not satisfying the thermal resistance required of the multilayer printed circuit board of a build up method also has it since the glass fiber base material is not used for layer insulation adhesives, as a solution of these points, it is already indicated on the Japanese-Patent-Application-No. No. 225235 [07 to] description, and the Japanese-Patent-Application-No. No. 194221 [09 to] descriptions. However, the halogenide is used for flameproofing and former invention is requiring not the thing that satisfies the safety in an environmental side but the matter which does not use a halogenide on environmental sanitation again.
[0005]
[Problem(s) to be Solved by the Invention] The copper foil which this invention was examined in order to solve the problem by the simplified build up method to the approach of fabricating with the above-mentioned hot-platen press, and it was completed, and was excellent in fire retardancy, high thermal resistance, a high electrical property, and preservation stability with especially the non halogen, and coated the layer insulation adhesives for epoxy resin system multilayer printed wiring boards and it 100 degrees C or more which can be promptly hardened at an elevated temperature is offered.
[0006]
[Means for Solving the Problem] This invention is copper foil which comes to coat the layer insulation adhesives for multilayer printed wiring boards and it which are characterized by containing each following component as an indispensable component. (b) the sulfur component content thermoplastics of weight average molecular weight 103-105, and (**) -- a 500 or less inorganic filler (Ha) weight per epoxy equivalent epoxy resin and a (d) epoxy resin curing agent, and [0007] In this invention, it is thought that it also raises an electrical property further although the sulfur component content thermoplastics of the weight average molecular weight 103-105 of a (b) component is blended for the object of making small flameproofing in the non halogen of an epoxy resin constituent and resin softening at the time of shaping, and maintaining the thickness of an insulating layer, giving flexibility to an epoxy resin constituent, and a raise in the heatproof of insulating resin. (b) As sulfur component content thermoplastics of the weight average molecular weight 103-105 of a component, they are polyphenylene sulfide, Pori Sall John, a polyether ape phone, and bisphenol smooth S form phenoxy resin. The addition rate of this amount sulfur component content thermoplastics of macromolecules is 30 - 90 % of the weight to the whole resin which is sum total weight with the epoxy resin of a component (Ha). If there are few additions than 30 % of the weight, flameproofing cannot

fully be discovered, but on the other hand, if [than 90 % of the weight] more, although flameproofing can be discovered, since an adhesives constituent lacks in resiliency firmly, the flattery nature to the irregularity of the base material at the time of lamination shaping and adhesion will be bad, and will cause shaping void generating. Moreover, while the end of this sulfur component content thermoplastics will stop the phase separation of sulfur component content thermoplastics and an epoxy resin after heat curing since reactivity with an epoxy resin is also good if a hydroxyl group, a carboxyl group, or amino-group denaturation is performed, in order for the thermal resistance of a hardened material to also improve, it is desirable to perform the above-mentioned denaturation.

[0008] In order to carry out a coat to that it is smeared at the time of a roll lamination, and a sex and adhesion are missing, that the adhesive property after a lamination is not enough, and copper foil, when it dissolves in a solvent and considers as the varnish of predetermined temperature, viscosity is high, it is smeared at the time of a coat and a sex and workability are not [the above-mentioned sulfur component content thermoplastics independent] good. In order to improve such a fault, a 500 or less weight per epoxy equivalent [which is a component (Ha)] epoxy resin is blended. This blending ratio of coal is 10 - 70% of the weight of the whole resin. The above-mentioned effectiveness is not expectable, and when 70 % of the weight is exceeded, it becomes impossible to expect the effectiveness of said amount sulfur component content thermoplastics of macromolecules at less than 10 % of the weight.

[0009] (c) Although there are the bisphenol A mold epoxy resin, a bisphenol female mold epoxy resin, a phenol novolak mold epoxy resin, a cresol novolak mold epoxy resin, and an aminophenol mold epoxy resin as an epoxy resin of a component, if the thing containing hetero atoms, such as a novolak mold epoxy resin, sulfur, and nitrogen, is used for fire-resistant grant, flameproofing of a multilayer printed wiring board will be performed more effectively.

[0010] (b) As an inorganic filler of a component, it is fused silica, a crystalline silica, a calcium carbonate, an aluminum hydroxide, an alumina, a magnesium hydroxide, clay, a barium sulfate, a mica, talc, white carbon, E glass impalpable powder, etc., and blend five to 50% of the weight to a component (Ha). If it blends more mostly than 50 % of the weight, the viscosity of adhesives will become high and the embedded nature of a between [inner layer circuits] will come to fall. The reduction in coefficient of linear expansion and heat-resistant improvement are expected by these combination.

[0011] Next, although especially a (d) epoxy resin curing agent is not limited [acid anhydride / an amine compound, an imidazole compound,], since loadings can fully stiffen an epoxy resin at least, an imidazole compound has them. [desirable] The solubility to an epoxy resin is small, it is a solid in ordinary temperature with a melting point of 130 degrees C or more, and an epoxy resin and especially its thing that reacts promptly are [an imidazole compound becomes an elevated temperature 150 degrees C or more, and] desirable. Specifically, there is 2-methylimidazole, 2-phenylimidazole, 2-phenyl-4-methylimidazole, a screw (2-ethyl-4-methylimidazole), 2-phenyl-4-methyl-5-hydroxymethylimidazole, 2-phenyl-4, 5-dihydroxymethylimidazole, or a triazine addition mold imidazole. These imidazoles are distributed by homogeneity in

an epoxy resin varnish as impalpable powder. Since compatibility with an epoxy resin is small, at ordinary temperature -100 degree C, a reaction does not advance, therefore preservation stability can be kept good. And if it heats at 150 degrees C or more at the time of lamination hardening, it will react with an epoxy resin and a uniform hardened material will be obtained.

[0012] As a curing agent, in addition, phthalic anhydride, an anhydrous tetrahydrophtal acid, an anhydrous methyl tetrahydrophtal acid, Methyl endo-methylene-tetrahydrophthalic anhydride, an anhydrous methyl butenyl tetrahydrophtal acid, Anhydrous hexahydrophthalic acid, anhydrous methyl hexahydrophthalic acid, anhydrous hexahydrophthalic acid, Acid anhydrides, such as trimellitic anhydride, pyromellitic dianhydride, and anhydrous benzophenone tetracarboxylic acid, the amine complex of a boron trifluoride, a dicyandiamide, or its derivative is mentioned, and what adduct[epoxy]-ized these, and the thing which microencapsulated can also be used. However, in case these curing agents are used, in order to obtain what carried out full hardening more, addition of the basic hardening accelerator usually used is needed for the inside of a short time.

[0013] The component which reacts can be blended with the epoxy resin and curing agent other than the above-mentioned epoxy resin and a curing agent. For example, they are epoxy reactivity diluents (glycerol triglycidyl ether as 3 organic-functions molds, such as resorcinol diglycidyl ether and ethylene glycol glycidyl ether, as 2 organic-functions molds, such as phenyl glycidyl ether, as 1 organic-functions mold etc.), a resol mold or novolak mold phenol system resin, an isocyanate compound, etc. furthermore, the defoaming agent for preventing silane coupling agents, such as an epoxy silane, or a titanate system coupling agent, and a void, in order to heighten the adhesion force with copper foil or the inner layer circuit board or to raise moisture resistance -- or addition of a liquefied or impalpable powder type flame retarder is also possible.

[0014] After applying adhesives to copper foil and drying as a solvent to be used, what does not remain into adhesives must be chosen. For example, an acetone, a methyl ethyl ketone (MEK), toluene, a xylene, n-hexane, a methanol, ethanol, methyl Cellosolve, ethyl Cellosolve, a cyclohexanone, dimethyl formamide (DMF), etc. are used.

[0015] Coating of the adhesives varnish which dissolved the adhesives component in the predetermined solvent by predetermined concentration is carried out to the support side of copper foil, the copper foil with layer insulation adhesives performs 80 degrees C - 130 degrees C desiccation after that, and in adhesives, as a solvent does not remain, it produces it. The thickness of the adhesives layer has 15 micrometers - desirable 120 micrometers. Although layer insulation nature is satisfactory if thicker [when thinner than 15 micrometers, layer insulation nature may become inadequate and] than 120 micrometers, it stops suiting the object of this invention that production is not easy and makes thickness of a multilayer board thin. This copper foil with layer insulation adhesives is usually laminated in the inner layer circuit board with a dry film laminator, can be stiffened, and can form the multilayer printed wiring board which has an outer layer circuit easily.

[0016] Next, the under coat agent used in order to lose the level difference by the circuit of

the inner layer circuit board is described. In order to usually layer insulation adhesives and really stiffen an under coat agent, this and an ingredient of the same kind are used. Therefore, what uses an epoxy resin as a principal component in this invention is used. However, the varnish which dissolved in the solvent is sufficient and the varnish which dissolved in the reactant diluent which reacts by heat or light is sufficient. this under coat agent varnish -- a inner layer circuit plate -- applying -- subsequently -- heating -- evaporation or the reaction of a solvent -- the formation of tuck free thru/or prepolymer-izing, or the formation of tuck free carry out an optical exposure and according to a reaction -- or it prepolymer-izes.

[0017]

[Example]

<<example 1>> The end hydroxyl-group denaturation polyether ape phone [average molecular weight 24000, Sumitomo Chemical Co., Ltd. make 5003P] 100 weight section, Weight per epoxy equivalent 190, the phenol novolak mold epoxy resin [Epiclon N-770 made from Dainippon Ink Chemistry] 30 weight section, The bisphenol female mold epoxy resin [weight-per-epoxy-equivalent 175 and Epiclon 830made from Dainippon Ink Chemistry-S] 15 weight section MEK, It stirred and dissolved at the DMF mixed solvent, the 2-methylimidazole 5 weight section, the titanate system coupling agent [KR[by Ajinomoto Co., Inc.]-46B] 0.2 weight section, and the barium-sulfate 20 weight section were added as a curing agent there, and the adhesives varnish was produced. Hereafter, the multilayer printed wiring board was produced at the process shown in drawing 1 . It applied and dried with the roller coater and copper foil with adhesives (3) was obtained so that the thickness after drying said adhesives varnish to the support side of copper foil (1) with a thickness of 18 micrometers might be set to 50 micrometers (a). Next, the bisphenol A mold epoxy resin (weight-per-epoxy-equivalent 470, weight average molecular weight 900 [about]) 100 weight section was dissolved in the glycidyl methacrylate 40 weight section, the 2-methylimidazole 3 weight section and the photopolymerization initiator (Ciba-Geigy IRUGA cure 651) 1.2 weight section were added as a curing agent to this, and it stirred enough in the homomixer, and considered as the under coat agent. Furthermore, pattern processing of the glass epoxy double-sided copper clad laminate of 0.1mm of base material thickness and 35 micrometers of copper foil thickness was carried out, and the inner layer circuit plate was obtained. a copper foil front face -- melanism -- after processing, coating of the above-mentioned under coat agent was carried out to about 40 micrometers in thickness by the curtain coating machine. Then, they are about 2 J/cm2 with two 80 W/cm high-pressure mercury-vapor lamps at UV conveyor. UV irradiation was carried out on conditions and the under coat agent was made tuck free. It is the above-mentioned copper foil with layer insulation adhesives on the inner layer circuit plate which has the layer of this under coat agent The temperature of 100 degrees C, and pressure 4 kg/cm2 From the conditions for lamination speed 0.8m/, the above-mentioned copper foil with heat-curing mold insulation adhesives was laminated using the hard roll, heat hardening was carried out for 30 minutes, and 150 degrees C of multilayer printed wiring boards were produced.

[0018] <<example 2>> The end hydroxyl-group denaturation polyether ape phone

[average molecular weight 24000, Sumitomo Chemical Co., Ltd. make 5003P]100 weight section, Weight per epoxy equivalent 190, the phenol novolak mold epoxy resin [Epiclon N-770 made from Dainippon Ink Chemistry] 70 weight section, Weight per epoxy equivalent 175 and the bisphenol female mold epoxy resin [Epiclon 830 made from Dainippon Ink Chemistry] 30 weight section MEK, It stirred and dissolved at the DMF mixed solvent, the 2-methylimidazole 5 weight section, the titanate system coupling agent [KR[by Ajinomoto Co., Inc.]-46B] 0.2 weight section, and the barium-sulfate 20 weight section were added as a curing agent there, and the adhesives varnish was produced. About multilayer printed wiring board production, it carried out like the example 1.

[0019] <<example 3>> The product made from Pori Sall John [Teijin Amoco Engineering plastics, The YUDERU P-1700]100 weight section, the phenol novolak mold epoxy resin [weight per epoxy equivalent 285, Nippon Kayaku Co., Ltd. make BREN-S] 40 weight sections, Weight per epoxy equivalent 175 and the bisphenol female mold epoxy resin [Epiclon 830 made from Dainippon Ink Chemistry] 20 weight section are stirred and dissolved at MEK. As the methyl-cyclohexene-dicarboxylic-anhydride 35 weight section and a hardening accelerator as a curing agent there The 2-phenyl-4-methyl-5-hydroxymethylimidazole 0.5 weight section, The titanate system coupling agent [KR[by Ajinomoto Co., Inc.]-46B] 0.2 weight section and the barium-sulfate 20 weight section were added, the adhesives varnish was produced, and the multilayer printed wiring board was produced like the example 1.

[0020] <<example 4>> The end hydroxyl-group denaturation polyether ape phone [average molecular weight 24000, Sumitomo Chemical Co., Ltd. make 5003P]100 weight section, Weight per epoxy equivalent 190, the phenol novolak mold epoxy resin [Epiclon N-770 made from Dainippon Ink Chemistry] 30 weight section, The bisphenol female mold epoxy resin [weight-per-epoxy-equivalent 175 and Epiclon 830made from Dainippon Ink Chemistry-S] 15 weight section MEK, It stirred and dissolved at the DMF mixed solvent, and the 2-methylimidazole 5 weight section and the magnesium-hydroxide 30 weight section were added as a curing agent there, the adhesives varnish was produced, and the multilayer printed wiring board was produced like the example 1.

[0021] Example of <<comparison 1>> The multilayer printed wiring board was obtained like the example 1 except having used the bisphenol A mold phenoxy resin [mean-molecular-weight 30000] 100 weight section, and weight per epoxy equivalent 175 and the bisphenol female mold epoxy resin [Epiclon 830 made from Dainippon Ink Chemistry] 40 weight section.

Example of <<comparison 2>> The end hydroxyl-group denaturation polyether ape phone [average molecular weight 24000, Sumitomo Chemical Co., Ltd. make 5003P]100 weight section, Weight per epoxy equivalent 190, the phenol novolak mold epoxy resin [Epiclon N-770 made from Dainippon Ink Chemistry] 80 weight section, Weight per epoxy equivalent 175 and the bisphenol female mold epoxy resin [Epiclon 830 made from Dainippon Ink Chemistry] 40 weight section are used. The multilayer printed wiring board was produced like the example 1 except having removed the titanate system coupling agent [KR[by Ajinomoto Co., Inc.]-46B], and the barium sulfate.

[0022] About the multilayer printed wiring board obtained above, surface smooth nature,

moisture absorption solder thermal resistance, the Peel reinforcement, and fire retardancy were measured, and the result shown in a table 1 was obtained.
<<measuring method>>
inner layer circuit plate test piece: -- 150-micrometer pitch between lines, and clearance hole 1.0mm -- phi1. surface smooth nature:JIS B 0601 R (max)
2. Moisture absorption solder thermal-resistance moisture absorption conditions : the number to which five test pieces blistered in 280 degrees C and 120 seconds was seen by pressure cooker processing, 125 degrees C, 2.3 atmospheric pressures, and 30 minute test condition:n=5.
3. Peel Reinforcement : JIS C 4. Fire Retardancy by 6486: JIS C [0023] by 6481
Table 1 Surface smooth nature Moisture absorption solder Peel reinforcement Fire retardancy (micrometer) Thermal resistance (kg/cm) An example 1 5 0 1.4 V-0 Example 2 3 0 1.3 V-0 Example 3 5 0 1.3 V-0 Example 4 5 0 1.3 V-0 Example 1 of a comparison 5 0 1.3 Combustion Example of comparison 2 3 3 1.4 Combustion [0024]
[Effect of the Invention] Since hardening is really performed good when laminated in the inner layer circuit board to which the layer insulation adhesives for multilayer printed wiring boards of this invention were excellent in shelf life in the condition of having carried out the coat to the condition or copper foil of a varnish, and coating of the under coat agent was carried out, especially the obtained multilayer printed wiring board is excellent in fire retardancy, an environmental side, and thermal resistance, and has the property which was excellent also in moisture resistance etc. not to mention the electrical property.

[Translation done.]

CLAIMS

[Claim(s)]
[Claim 1] Layer insulation adhesives for multilayer printed wiring boards characterized by containing each following component as an indispensable component.
(b) the sulfur component content thermoplastics of weight average molecular weight 103-105, and (**) -- an inorganic filler, a 500 or less weight per epoxy equivalent (Ha) epoxy resin and a (d) epoxy resin curing agent, and [Claim 2] (b) a component -- (**) -- the layer insulation adhesives for multilayer printed wiring boards according to claim 1 which are 30 - 90% of the weight of the sum total weight of a component and (Ha) a component.
[Claim 3] (b) Layer insulation adhesives for multilayer printed wiring boards according to claim 1 or 2 whose component is 5 - 50% of the weight of a component (Ha).
[Claim 4] (b) Layer insulation adhesives for multilayer printed wiring boards according to claim 1, 2, or 3 whose sulfur component content thermoplastics of the weight average molecular weight 103-105 of a component is polyphenylene sulfide, Pori Sall John, a polyether ape phone, and bisphenol smooth S form phenoxy resin.
[Claim 5] (c) Layer insulation adhesives for multilayer printed wiring boards according to claim 1, 2, 3, or 4 whose component is one sort chosen from the bisphenol mold epoxy resin, the novolak mold epoxy resin, and the aminophenol mold epoxy resin, or two sorts or more.
[Claim 6] Copper foil for multilayer printed wiring boards which comes to coat layer insulation adhesives according to claim 1, 2, 3, 4, or 5 copper foil.

[Translation done.]

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(54)【発明の名称】　多層プリント配線板用層間絶縁接着剤及び接着

(57)【要約】
【課題】　ノンハロゲンで難燃性を示し、耐熱性、保存安定性に優れ、かつ100℃以上の高温で速やかに硬化しうるエポキシ樹脂系層間絶縁接着剤を得ること。
【解決手段】　下記の各成分を必須成分として含有する多層プリント配線板用層間絶縁接着剤。
（1）重量平均分子量10^3～10^5の硫黄成分含有熱可塑性樹脂、（2）無機充填材、（3）エポキシ当量500以下のエポキシ樹脂、及び（4）エポキシ樹脂硬化剤

【特許請求の範囲】

【請求項1】　下記の各成分を必須成分として含有することを特徴とする多層プリント配線板用層間絶縁接着剤.

(イ) 重量平均分子量10'～10'の硫黄成分含有熱可塑性樹脂、(ロ) 無機充填材、(ハ) エポキシ当量500以下のエポキシ樹脂、及び(ニ) エポキシ樹脂硬化剤.

【請求項2】　(イ) 成分が、(イ) 成分及び(ハ) 成分の合計重量の30～90重量%である請求項1記載の多層プリント配線板用層間絶縁接着剤.

【請求項3】　(ロ) 成分が、(ハ) 成分の5～50重量%である請求項1又は2記載の多層プリント配線板用層間絶縁接着剤.

【請求項4】　(イ) 成分の重量平均分子量10'～10'の硫黄成分含有熱可塑性樹脂が、ポリフェニレンサルファイド、ポリサルフォン、ポリエーテルサルフォン、ビスフェノールS型フェノキシ樹脂である請求項1、2又は3記載の多層プリント配線板用層間絶縁接着剤.

【請求項5】　(ハ) 成分が、ビスフェノール型エポキシ樹脂、ノボラック型エポキシ樹脂、及びアミノフェノール型エポキシ樹脂から選ばれた1種又は2種以上である請求項1、2、3又は4記載の多層プリント配線板用層間絶縁接着剤.

【請求項6】　請求項1、2、3、4又は5記載の層間絶縁接着剤を銅箔にコーティングしてなる多層プリント配線板用銅箔.

【発明の詳細な説明】

【0001】

【発明の属する技術分野】本発明は、エポキシ樹脂系多層プリント配線板用層間絶縁接着剤及びそれをコーティングした銅箔に関するものである.

【0002】

【従来の技術】従来、多層プリント配線板を製造する場合、回路が形成された内層回路基板上にガラスクロス基材にエポキシ樹脂を含浸して半硬化させたプリプレグシートを1枚以上重ね、更にその上に銅箔を重ね熱板プレスにて加圧一体成形するという工程を経ている。しかし、この工程ではプリプレグ中の含浸樹脂を熱により再流動させ一定圧力下で硬化させるため、均一に硬化成形させるためには1～1.5時間は必要である。このように製造工程が長くかかる上に、多層積層プレス及びガラスクロスプリプレグのコスト等により高コストとなっている。加えてガラスクロスに樹脂を含浸させる方法のため、回路層間の厚みがガラスクロスにより制限され多層プリント配線板全体の極薄化も困難であった.

【0003】近年、これらの問題を解決するため、熱板プレスによる加熱加圧成形を行わず、層間絶縁材にガラスクロスを用いない、ビルドアップ方式による多層プリント配線板の技術が改めて注目されている。ビルドアップ方式による多層プリント配線板において、フィルム状の層間絶縁樹脂層を用いた場合、内層回路板の絶縁基板と回路と段差を無くし、その表面を平滑化するために、内層回路板にアンダーコート剤を塗布することが一般化してきた。この代表的な例として、内層回路板に塗布されたアンダーコート剤が未硬化、半硬化または硬化した状態において、層間絶縁接着剤をコートした銅箔をラミネートし、一体硬化することにより多層プリント配線板を得る。このような方法により、内層回路板の回路による段差が小さくなるため、層間絶縁接着剤をコートした銅箔のラミネートが容易であり、また内層回路板の銅箔残存率を考慮する必要も少なくなる.

【0004】このようなプロセスにおいて、銅箔にコートされた層間絶縁接着剤がラミネート成形時に軟化し過ぎて層間絶縁厚みを確保できない。熱硬化時に溶融粘度が下がり過ぎて銅が発生する。また、その保存時に硬化反応が進行して、アンダーコート剤が塗布された内層回路板にラミネートしたとき一体成形が良好に行われないと言う問題が生じている。更に、層間絶縁接着剤にガラス繊維基材が使用されていないため、難燃化が困難であり、多層プリント配線板の高密度化に伴う、ビルドアップ方式の多層プリント回路板に要求される耐熱性を満足しないという問題点もあるが、これらの点の解決策としては既に特願平07-225235号明細書、特願平09-194221号明細書で開示されている。しかし、以前の発明では難燃化のためにハロゲン化物を使用しており、環境面における安全性を満足するものではなく、また環境衛生上、ハロゲン化物を用いない物質が要求されつつある.

【0005】

【発明が解決しようとする課題】本発明は、上記熱板プレスで成形する方法に対して、簡素化されたビルドアップ方式による問題を改善するために検討し、完成されたものであり、特にノンハロゲンで難燃性、高耐熱性、高靭気特性、保存安定性に優れ、かつ100℃以上の高温で速やかに硬化し得るエポキシ樹脂系多層プリント配線板用層間絶縁接着剤及びそれをコーティングした銅箔を提供するものである.

【0006】

【課題を解決するための手段】本発明は、下記の各成分を必須成分として含有することを特徴とする多層プリント配線板用層間絶縁接着剤及びそれをコーティングしてなる銅箔である.

(イ) 重量平均分子量10^3～10^5の硫黄成分含有熱可塑性樹脂、(ロ) 無機充填材

(ハ) エポキシ当量500以下のエポキシ樹脂、及び

(ニ) エポキシ樹脂硬化剤.

【0007】本発明において、(イ) 成分の重量平均分子量10^3～10^5の硫黄成分含有熱可塑性樹脂は、エ

ポキシ樹脂組成物のノンハロゲンでの難燃化、成形時の樹脂軟化を小さくし、絶縁層の厚みを維持すること、エポキシ樹脂組成物に可撓性を付与すること、絶縁樹脂の高耐熱化の目的で配合されているが、更に、電気特性をも向上させると考えられる。（イ）成分の重量平均分子量10^3～10^5の硫黄成分含有熱可塑性樹脂としては、ポリフェニレンサルファイド、ポリサルフォン、ポリエーテルサルフォン、ビスフェノールS型フェノキシ樹脂である。この高分子量硫黄成分含有熱可塑性樹脂の添加割合は（ハ）成分のエポキシ樹脂との合計重量である樹脂全体に対して30～90重量%である。添加量が30重量%より少ないと、難燃化を十分に発現できず、一方、90重量%より多いと、難燃化は発現できるが、接着剤組成物が硬く弾力性に欠けるため、ラミネート成形時の基材の凹凸への追従性、密着性が悪く、成形ボイド発生の原因となる。また、この硫黄成分含有熱可塑性樹脂の末端が水酸基、カルボキシル基、あるいはアミノ基変性が行われておれば、エポキシ樹脂との反応性も良いことから熱硬化後に硫黄成分含有熱可塑性樹脂とエポキシ樹脂との相分離を抑えるとともに、硬化物の耐熱性も向上するため上記変性が行われていることが望ましい。

【0008】上記硫黄成分含有熱可塑性樹脂単独では、ロールラミネート時の塗れ性、密着性に欠けること、ラミネート後の接着性が十分でないこと、及び銅箔にコートするために溶剤に溶解して所定濃度のワニスとしたときに、粘度が高く、コート時の塗れ性、作業性が良くない。このような欠点を改善するために（ハ）成分であるエポキシ当量500以下のエポキシ樹脂を配合する。この配合割合は樹脂全体の10～70重量%である。10重量%未満では上記の効果が期待できず、また、70重量%を越えると前記高分子量硫黄成分含有熱可塑性樹脂の効果が期待できなくなる。

【0009】（ハ）成分のエポキシ樹脂としてはビスフェノールA型エポキシ樹脂、ビスフェノールF型エポキシ樹脂、フェノールノボラック型エポキシ樹脂、クレゾールノボラック型エポキシ樹脂、アミノフェノール型エポキシ樹脂があるが、難燃性付与のためにはノボラック型エポキシ樹脂、硫黄、窒素などのヘテロ原子を含むものを使用すれば、多層プリント配線板の難燃化がより効果的に行われる。

【0010】（ロ）成分の無機充填材としては、溶融シリカ、結晶性シリカ、炭酸カルシウム、水酸化アルミニウム、アルミナ、水酸化マグネシウム、クレー、硫酸バリウム、マイカ、タルク、ホワイトカーボン、Eガラス微粉末などであり、（ハ）成分に対して5～50重量%配合する。50重量%より多く配合すると、接着剤の粘性が高くなり、内層回路間への埋込性が低下するようになる。これらの配合により低線膨張率化、耐熱性向上が期待される。

【0011】次に、（ニ）エポキシ樹脂硬化剤はアミン化合物、イミダゾール化合物、酸無水物など、特に限定されるものではないが、イミダゾール化合物は配合量が少なくてもエポキシ樹脂を十分に硬化させることができるので好ましいものである。イミダゾール化合物は、融点130℃以上の高温で固形であり、エポキシ樹脂への溶解性が小さく、150℃以上の高温になって、エポキシ樹脂と速やかに反応するものが特に好ましい。具体的には2-メチルイミダゾール、2-フェニルイミダゾール、2-フェニル-4-メチルイミダゾール、ビス（2-エチル-4-メチル-イミダゾール）、2-フェニル-4-メチル-5-ヒドロキシメチルイミダゾール、2-フェニル-4，5-ジヒドロキシメチルイミダゾール、あるいは、トリアジン付加型イミダゾール等がある。これらのイミダゾールは微粉末としてエポキシ樹脂ワニス中に均一に分散される。エポキシ樹脂との相溶性が小さいので、常温～100℃では反応が進行せず、従って保存安定性を良好に保つことができる。そしてラミネート硬化時に150℃以上に加熱すると、エポキシ樹脂と反応し、均一な硬化物が得られる。

【0012】その他硬化剤として、無水フタル酸、無水テトラヒドロフタル酸、無水メチルテトラヒドロフタル酸、無水メチルエンドメチレンテトラヒドロフタル酸、無水メチルブテニルテトラヒドロフタル酸、無水ヘキサヒドロフタル酸、無水メチルヘキサヒドロフタル酸、無水ヘキサヒドロフタル酸、無水トリメリット酸、無水ピロメリット酸、無水ベンゾフェノンテトラカルボン酸等の酸無水物、三フッ化ホウ素のアミン錯体、ジシアンジアミド又はその誘導体などが挙げられ、これらをエポキシアダクト化したものやマイクロカプセル化したものも使用できる。しかし、これら硬化剤を用いる際は、短時間のうちに、より完全硬化したものを得るために、通常使用される塩基性の硬化促進剤の添加を必要とする。

【0013】上記エポキシ樹脂及び硬化剤の他に、エポキシ樹脂や硬化剤と反応する成分を配合することができる。例えば、エポキシ反応性希釈剤（一官能型としてフェニルグリシジルエーテルなど、二官能型としてレゾルシンジグリシジルエーテル、エチレングリコールグリシジルエーテルなど、三官能型としてグリセロールトリグリシジルエーテルなど）、レゾール型又はノボラック型フェノール系樹脂、イソシアネート化合物などである。更に、銅箔や内層回路基板との密着力を高めたり、耐湿性を向上させるためにエポキシシラン等のシランカップリング剤あるいはチタネート系カップリング剤、ボイドを防ぐための消泡剤、あるいは液状又は微粉末タイプの難燃剤の添加も可能である。

【0014】使用する溶剤としては、接着剤を銅箔に塗布し乾燥した後において、接着剤中に残らないものを選択しなければならない。例えば、アセトン、メチルエチルケトン（MEK）、トルエン、キシレン、n-ヘキサ

ン、メタノール、エタノール、メチルセルソルブ、エチルセルソルブ、シクロヘキサノン、ジメチルフォルムアミド（DMF）などが用いられる。

【0015】層間絶縁接着剤付き銅箔は、接着剤成分を所定の溶剤に所定の濃度で溶解した接着剤ワニスを銅箔のアンカー面に塗工し、その後80℃～130℃の乾燥を行って接着剤中に溶剤が残らないようにして作製する。その接着剤層の厚みは15μm～120μmが好ましい。15μmより薄いと層間絶縁性が不十分となることがあり、120μmより厚いと層間絶縁性は問題ないが、作製が容易でなく、また多層板の厚みを薄くするという本発明の目的に合わなくなる。この層間絶縁接着剤付き銅箔は、通常ドライフィルムラミネーターにより内層回路基板にラミネートし硬化させて、容易に外層回路を有する多層プリント配線板を形成することができる。

【0016】次に、内層回路基板の回路による段差を無くすために用いられるアンダーコート剤について述べる。アンダーコート剤は通常層間絶縁接着剤と一体硬化させるために、これと同様の材料が使用される。従って、本発明においてはエポキシ樹脂を主成分とするものが使用される。ただし、溶剤に溶解したワニスでもよく、熱又は光により反応する反応性希釈剤に溶解したワニスでもよい。かかるアンダーコート剤ワニスを内層回路板に塗布し、次いで加熱して溶剤の蒸発あるいは反応によりタックフリー化ないしプレポリマー化、又は光照射して反応によるタックフリー化ないしプレポリマー化する。

【0017】

【実施例】

《実施例1》末端水酸基変性ポリエーテルサルフォン［平均分子量24000、住友化学工業（株）製　5003P］100重量部、フェノールノボラック型エポキシ樹脂［エポキシ当量190、大日本インキ化学（株）製　エピクロンN-770］30重量部、ビスフェノールF型エポキシ樹脂［エポキシ当量175、大日本インキ化学（株）製　エピクロン830-S］15重量部とをMEK、DMF混合溶媒に撹拌・溶解し、そこへ硬化剤として2-メチルイミダゾール5重量部、チタネート系カップリング剤［味の素（株）製　KR-46B］0.2重量部、硫酸バリウム20重量部を添加して接着剤ワニスを作製した。以下、図1に示す工程にて多層プリント配線板を作製した。前記接着剤ワニスを厚さ18μmの銅箔（1）のアンカー面に乾燥後の厚みが50μmとなるようにローラーコーターにて塗布、乾燥して接着剤付き銅箔（3）を得た（a）。次に、ビスフェノールA型エポキシ樹脂（エポキシ当量470、重量平均分子量約900）100重量部をグリシジルメタクリレート40重量部に溶解し、これに硬化剤として2-メチルイミダゾール3重量部と光重合開始剤（チバガイギー製イルガキュア651）1.2重量部を添加し、ホモミキサーにて十分撹拌してアンダーコート剤とした。更に、基材厚0.1mm、銅箔厚35μmのガラスエポキシ両面銅張積層板をパターン加工して内層回路板を得た。銅箔表面を黒化処理した後、上記アンダーコート剤をカーテンコーターにより厚さ約40μmに塗工した。その後、UVコンベア機にて80W/cm高圧水銀灯2本で約2J/cm²　の条件で紫外線照射し、アンダーコート剤をタックフリー化した。かかるアンダーコート剤の層を有する内層回路板上に上記層間絶縁接着剤付き銅箔を、温度100℃、圧力4Kg/cm²　、ラミネートスピード0.8m/分の条件より、硬質ロールを用いて上記熱硬化型絶縁性接着剤付き銅箔をラミネートし、150℃、30分間加熱硬化させ多層プリント配線板を作製した。

【0018】《実施例2》末端水酸基変性ポリエーテルサルフォン［平均分子量24000、住友化学工業（株）製　5003P］100重量部、フェノールノボラック型エポキシ樹脂［エポキシ当量190、大日本インキ化学（株）製　エピクロンN-770］70重量部、ビスフェノールF型エポキシ樹脂［エポキシ当量175、大日本インキ化学（株）製　エピクロン830］30重量部とをMEK、DMF混合溶媒に撹拌・溶解し、そこへ硬化剤として2-メチルイミダゾール5重量部、チタネート系カップリング剤［味の素（株）製　KR-46B］0.2重量部、硫酸バリウム20重量部を添加して接着剤ワニスを作製した。多層プリント配線板作製については実施例1と同様にして行った。

【0019】《実施例3》ポリサルフォン［帝人アモコエンジニアリングプラスチックス（株）製、ユーデルP-1700］100重量部、フェノールノボラック型エポキシ樹脂［エポキシ当量285、日本化薬（株）製　BREN-S］40重量部、ビスフェノールF型エポキシ樹脂［エポキシ当量175、大日本インキ化学（株）製　エピクロン830］20重量部とをMEKに撹拌・溶解し、そこへ硬化剤としてメチルテトラヒドロ無水フタル酸35重量部、硬化促進剤として2-フェニル-4-メチル-5-ヒドロキシメチルイミダゾール0.5重量部、チタネート系カップリング剤［味の素（株）製　KR-46B］0.2重量部、硫酸バリウム20重量部を添加して接着剤ワニスを作製し、実施例1と同様にして多層プリント配線板を作製した。

【0020】《実施例4》末端水酸基変性ポリエーテルサルフォン［平均分子量24000、住友化学工業（株）製　5003P］100重量部、フェノールノボラック型エポキシ樹脂［エポキシ当量190、大日本インキ化学（株）製　エピクロンN-770］30重量部、ビスフェノールF型エポキシ樹脂［エポキシ当量175、大日本インキ化学（株）製　エピクロン830-S］15重量部とをMEK、DMF混合溶媒に撹拌・溶解し、そこへ硬化剤として2-メチルイミダゾール5重

量部、水酸化マグネシウム30重量部を添加して後者剤ワニスを作製し、実施例1と同様にして多層プリント配線板を作製した。

【0021】《比較例1》ビスフェノールA型フェノキシ樹脂［平均分子量30000］100重量部とビスフェノールF型エポキシ樹脂［エポキシ当量175、大日本インキ化学（株）製　エピクロン830］40重量部を使用した以外は実施例1と同様にして多層プリント配線板を得た。

《比較例2》末端水酸基変性ポリエーテルサルフォン［平均分子量24000、住友化学工業（株）製　5003P］100重量部、フェノールノボラック型エポキシ樹脂［エポキシ当量190、大日本インキ化学（株）製　エピクロンN-770］80重量部、ビスフェノールF型エポキシ樹脂［エポキシ当量175、大日本インキ化学（株）製　エピクロン830］40重量部とを使用し、チタネート系カップリング剤［味の素（株）製　＊KR-46B］及び、硫酸バリウムを除いた以外は実施例1と同様にして多層プリント配線板を作製した。

【0022】以上得られた多層プリント配線板について、表面平滑性、吸湿半田耐熱性、ピール強度及び難燃性を測定し、表1に示す結果を得た。

《測定方法》

内層回路板試験片：線間150μmピッチ、クリアランスホール1.0mmφ

1．表面平滑性：JIS B 0601 R（max）

2．吸湿半田耐熱性

吸湿条件：プレッシャークッカー処理、125℃、2.3気圧、30分

試験条件：n=5で、5個の試験片が280℃、120秒間で膨れた個数をみた。

3．ピール強度：JIS C 6486による

4．難燃性：　JIS C 6481による

【0023】

表1

	表面平滑性（μm）	吸湿半田耐熱性	ピール強度（kg/cm）	難燃性
実施例1	5	0	1.4	V-0
実施例2	3	0	1.3	V-0
実施例3	5	0	1.3	V-0
実施例4	5	0	1.3	V-0
比較例1	5	0	1.3	燃焼
比較例2	3	3	1.4	燃焼

【0024】

【発明の効果】本発明の多層プリント配線板用層間絶縁接着剤は、ワニスの状態あるいは銅箔にコートした状態において、保存性にすぐれ、アンダーコート剤が塗工された内層回路基板にラミネートしたとき一体硬化が良好に行われるので、得られた多層プリント配線板は特に難燃性、環境面、耐熱性に優れ、電気特性はもちろんのこと、耐湿性等においても優れた特性を有している。

【図面の簡単な説明】

【図1】本発明の層間絶縁接着剤を用いて多層プリント配線板を作製する工程の一例を示す概略断面図

【符号の説明】

1　内層回路板
2　内層回路
3　アンダーコート剤
4　熱硬化型絶縁接着剤
5　銅箔
6　硬質ロール
7　多層プリント配線板